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## MEMORANDUM REPORT BRL-MR-3440

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# IMBEDDED THERMOCOUPLES AS A SOLID PROPELLANT COMBUSTION PROBE

Martin S. Miller  
Terence P. Coffee  
Anthony J. Kotlar

April 1985

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undertaken in order to evaluate sources of error involved and to make an assessment of the potential for success in deriving overall kinetics from such a measurement. The results of this review along with original calculations of condensed phase response errors are reported herein.

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## I. INTRODUCTION

Attempts to understand the combustion mechanisms of solid propellants have led to the development of a number of mathematical models which use simplified descriptions of reactive energy release in the condensed and gas phases. In each phase, what are likely to be complex reaction networks are treated as single formal reactions so as to circumvent the uncertainties that exist over their actual chemical nature while preserving generalized pressure and temperature dependences typical of such reactions. It has become customary to determine the effective kinetics of these formal reactions by adjusting them to give best fits of the model to burning rates as functions of pressure (and sometimes initial temperature). Such a procedure, of course, presumes that the overall reaction description is adequate for each phase, that the effective kinetics parameters do not change with pressure (or initial temperature), and that these parameters may be reliably extracted from the combustion model. The adequacy of the formal reaction description in the solid phase and its connection with burning rate (i.e., the usual pyrolysis law) remains as an important research task. In the gas phase, recent work<sup>1,2</sup> suggests that the single reaction formalism is sound but that the effective kinetics parameters for gas phase reactions can vary considerably with pressure. The reliability with which the kinetics parameters may be extracted from burning rate data alone has also been called into question.<sup>3,4</sup> Clearly, what is required is a means of characterizing the rate of energy release independently for each phase. Such means has been developed<sup>5,6</sup> but requires measurement of the temperature profile through the combustion wave.

At present the only technique which has been used to map temperature profiles through the entire combustion wave of solid propellants is that of imbedded microthermocouples. Various laser raman techniques are being developed to provide non-intrusive temperature measurements in the gaseous

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<sup>1</sup>T.P. Coffee, A.J. Kotlar, and M.S. Miller, "The Overall Reaction Concept in Premixed, Laminar, Steady-State Flames. I. Stoichiometries, Combustion & Flame, Vol. 54, pp. 155-169, 1983.

<sup>2</sup>T.P. Coffee, A.J. Kotlar, and M.S. Miller, "The Overall Reaction Concept in Premixed, Laminar, Steady-State Flames. II. Pressure and Initial Temperature," to be published in Combustion & Flame.

<sup>3</sup>M.S. Miller and T.P. Coffee, "A Fresh Look at the Classical Approach to Homogeneous Solid Propellant Combustion Modeling," Combustion & Flame, Vol. 50, pp. 65-74, 1983.

<sup>4</sup>M.S. Miller and T.P. Coffee, "On the Numerical Accuracy of Homogeneous Solid Propellant Combustion Models," Combustion & Flame, Vol. 50, pp. 75-88, 1983.

<sup>5</sup>A.A. Zenin, "Formal Kinetic Characteristics of the Reactions Accompanying the Burning of a Powder," Fizika Goreniya i Vzryva, Vol. 2, pp. 28-32, 1966.

<sup>6</sup>A.J. Kotlar, M.S. Miller, and T.P. Coffee, "Effective Kinetic Parameters for Gas Phase Heat Release During Solid Propellant Combustion," Proceedings of the 21st JANNAF Combustion Meeting, October 1984.

combustion zones but their spatial resolution at present is insufficient to probe fizz zone details and thermocouples may remain the best method in the condensed phase for the foreseeable future.

The imbedded thermocouple technique is hardly new, having been first reported in 1950<sup>7</sup> and practiced by some dozen groups since then; yet questions as to its accuracy and limitations persist. Because of the renewed importance of temperature data to combustion modeling, the present study was undertaken to assess the reliability of previous measurements of this type and to determine if refinements might profitably be attempted.

Systematic errors in imbedded thermocouple measurements can arise from a number of sources. These include experimental details relating to the assurance of one dimensional burning, mechanical stability of the thermocouple leads, heat conduction through the leads, radiation loss from the junction, catalytic effects, and junction "response." In the discussion to follow, we will review what has been learned of these problems in previous work, extend these analyses where possible, and point out what further work might assist in defining the limitations of the method.

## II. SOURCES OF ERROR

### A. Conditions For 1-D Burning

A great many experimental factors potentially affect the meaningfulness of temperature profiles obtained with imbedded thermocouples. Of basic importance is the assurance of reproducible, one-dimensional, and steady-state combustion of the propellant sample. Achievement of this condition is probably most closely approached with a cylindrical strand of propellant burned in a vented chamber. A shroud of inert gas flows continuously over the burning sample, the total pressure being maintained at a constant value using either a regulator on the inert gas supply or a back-pressure regulator on the chamber. The lateral surface of the sample is usually inhibited with coatings of materials less flammable than the propellant to keep the flame from spreading down the sides. Care is required to insure that the measured burning rate is independent of the coating type or thickness, the inert gas flow rate, and the diameter of the strand. Although more sophisticated techniques may be more convenient, the recorded burn-through of spaced fuse wires can provide adequate precision<sup>8</sup> (.33%) in measurements of the linear burning rate.

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<sup>7</sup>R. Klein, M. Mentser, G. von Elbe, and B. Lewis, "Determination of the Thermal Structure of a Combustion Wave by Fine Thermocouples," J. Physical and Colloid Chem., Vol. 54, pp 877-884, 1950.

<sup>8</sup>B.L. Crawford, Jr. and Clayton Huggett, "Direct Measurement of Burning Rates by an Electric Timing Method," National Defense Research Committee Report No. A-286, August 1944.



## B. Thermal Expansion Of The Leads

A potential source of error, which appears not to have been examined previously, is that caused by thermal expansion of the thermocouple leads. Such elongation makes the idealized linear relationship between time and distance only approximately correct. (Temperature vs. time is recorded; temperature vs. distance is the desired quantity.) We have analyzed this problem as follows.

To maximize the effect, we assume that the thermal expansion process occurs instantaneously. Then, at some particular instant, a wire extended along a normal to the surface into the fizz zone will have its differential elements expanded to a degree depending on its local temperature. If  $d\ell$  is the expanded element length at a distance  $\ell$  from the surface and  $dx$  is the length of this element at the surface temperature  $T_s$ , then

$$d\ell = dx + \alpha[T(\ell) - T_s] dx \quad (1)$$

where  $\alpha$  is the linear expansion coefficient. Eq. 1 is valid for small fractional elongations. Integrating this equation to a distance  $\ell$  from the surface

$$\ell = \alpha \int_0^\ell \frac{T(\ell')}{\left(\frac{d\ell'}{dx}\right)} d\ell' + (1 - \alpha T_s)x \quad (2)$$

where  $x$  would be the corresponding length if the fizz zone were at constant temperature  $T_s$ . We can avoid solving this integro-differential equation, by observing that  $\frac{d\ell}{dx} > 1$  and is monotonically increasing for a monotonically increasing temperature profile. Then setting  $\frac{d\ell}{dx} = 1$  in Eq. 2 will result in a simple integral equation which will yield an upper bound for  $\ell$ . We have solved this resulting equation using an analytic approximation to the temperature profile measured by Kubota<sup>9</sup> at 21 atm. The maximum elongation of only one micron occurs at the end of the fizz zone. The maximum temperature error, i.e.,  $T(\ell) - T(x)$ , occurs near the end of the fizz zone and is no greater than 2.5°C. Unless steeper gradients or higher temperatures are encountered, this source of error may safely be disregarded.

## C. Heat Conduction Through The Leads

Although many variations have been tried, the thermocouple is typically imbedded<sup>7</sup> by splitting a cylindrical strand along a plane through its axis, wetting the surface with propellant solvent, sandwiching the thermocouple between the strand halves, then drying under heat and light pressure. The manner in which the thermocouple wires are led away from the junction and out through the sides of the strand may be a critical determinant in the ultimate

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<sup>9</sup>N. Kubota, T.J. Ohlemiller, L.H. Caveny, and M. Summerfield, "The Mechanism of Super-Rate Burning of Catalyzed Double Base Propellants," AMS Report No. 1087, Princeton University, March 1973.

accuracy of the technique. To reduce errors due to thermal conduction through these wires, it would seem best to arrange them parallel to the burning surface for a short distance before angling them toward the cooler end of the strand. However, as pointed out by Strittmater, et al.,<sup>10</sup> the temperature gradient at the surface is so high ( $10^{\circ}\text{C}/\mu\text{m}$ ) that the burning surface may reach the leads before the bead (since one cannot expect the surface to burn with perfect planarity normal to the strand axis). Such a lead orientation would also be particularly sensitive to vibration induced by the gases accelerating away from the surface. For these reasons the leads are generally oriented away from the surface at an acute angle to the strand axis. Unfortunately, the effect of heat conduction through the leads on temperature accuracy is difficult to analyze for this configuration.

Strittmater, et al.,<sup>10</sup> considered the leads as insulated conductors with exponential heat input into the hot end. Although some elements of this analysis are quite reasonable, the conclusion is reached that the conduction errors increase as the wire diameter decreases. In the limit of very thin wires, this is unlikely to be the case because the ratio of the surface area of the wire (per unit length) to the cross sectional area will increase as the diameter decreases. This allows each differential length element of the wire to accommodate to the surrounding propellant temperature with increasing efficiency. Thus, the assumption of insulated conductors is not a good one in the limit of small wire diameters.

A different approach was taken by Suh and Tsai,<sup>11</sup> who retained the complex geometry of the thermocouple and leads but transformed the problem to one dimension using the concept of heat transfer coefficients in a solid. The transient problem for the total response error in the solid was then solved numerically. Since there remained uncertainties in the heat transfer coefficients, they were "calibrated" by applying the analysis to a sequence of wire sizes for which experimental measurements were made. One could then determine actual temperatures by extrapolating the model to zero wire diameter. Their strategy was unique and valuable but probably demanded too much from the nascent concept of solid heat transfer coefficients, which were not appropriate to such large gradients.<sup>12</sup>

The problem of heat loss through the thermocouple leads has not yet yielded to analysis and probably requires solution of the full three-dimensional transient problem. However, given this degree of difficulty, one might be better advised to approach the matter experimentally by making systematic measurements with varying wire sizes and angles between the leads.

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<sup>10</sup>R.C. Strittmater, H.E. Holmes, and L.A. Watermeier, "Lead Loss Problem for a Thermocouple Imbedded in a Burning Propellant," J. of Spacecraft and Rockets, Vol. 3, pp. 1302-1303, 1966.

<sup>11</sup>N.P. Suh and C.L. Tsai, "Thermocouple Response Characteristics in Deflagrating Low-Conductivity Materials," Transactions of the ASME, J. of Heat Transfer, Vol. 93, pp. 77-87, 1971.

<sup>12</sup>See discussion and references following paper of Reference 11.



#### D. Surface Temperature

One key piece of information which is desired from temperature measurements of this kind is the value of the temperature at the burning surface of the propellant. Since the temperature profile through both condensed and gas phases is a continuous trace, one would like to identify the position of the surface on the profile. Four methods have been used to do this. The first<sup>7,9,13,14</sup> assumes that any reaction in the condensed phase takes place at or very close to the surface. Since the temperature profile in an ablating solid can be described analytically by an exponential function of distance, one simply examines the logarithm of the temperature profile as a function of distance and associates the surface with the first sign of departure from linearity of this curve. Since a rather arbitrary assumption is involved, this is the least satisfactory method. The second method<sup>15,16</sup> looks for a sudden jog in the temperature profile which might suggest a discontinuity either in the thermal conductivity or a sharp heat release at a phase boundary. Such a jog has sometimes but not always been noted. (Absence of this artifact could be caused by the integrating effect of too large a bead.) The third<sup>10,13,17</sup> method attempts to synchronize high speed cinematography with the temperature profile and note the point of emergence visually. The method appears to work for some clean-burning propellants but visual ambiguities often arise. Finally, Zenin<sup>15</sup> imbedded a third probe close to the thermocouple junction. He then monitored the electrical conductivity between this probe and one leg of the thermocouple. Since conductivity of the propellant increases with increasing temperature then falls to a low value in the gas phase, the peak in the conductivity curve is taken to indicate the location of the surface. Zenin also notes that this assignment coincides with a slight jog in the temperature trace, providing further confidence in the interpretation of this feature when present.

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<sup>13</sup>R.G. Nugent, R. Friedman, and K.E. Rumbel, "Temperature-Profile Studies in Solid-Propellant Flames," Atlantic Research Corp, Air Force Office of Scientific Research TN 57-212, March 1957, AD 126509.

<sup>14</sup>A.J. Sabadell, J. Wenograd, and M. Summerfield, "Measurement of Temperature Profiles through Solid-Propellant Flames Using Fine Thermocouples," AIAA J., Vol. 3, pp. 1580-1584, 1965.

<sup>15</sup>A.A. Zenin, "Structure of Temperature Distribution in Steady-State Burning of a Ballistite Powder," Fizika Goreniya i Vzryva, Vol. 2, pp. 67-76, 1966.

<sup>16</sup>A.A. Zenin and O.I. Nefedova, "Burning of Ballistite Powder over a Broad Range of Initial Temperatures," Fizika Goreniya i Vzryva, Vol. 3, pp. 45-53, 1967.

<sup>17</sup>N.P. Suh, C.L. Tsai, C.L. Thompson Jr., and J.S. Moore, "Ignition and Surface Temperatures of Double Base Propellants at Low Pressure: I. Thermocouple Measurements," AIAA J., Vol. 8, pp. 1314-1321, 1970.

## E. Catalytic Effects

A number of investigators have been sensitive to the possibility that the presence of the thermocouple material in the reactive gases near the burning surface could lead to local catalytic heating and hence to misleading temperatures. Klein, et al.,<sup>7</sup> analyzed the fizz zone gases and found NO, H<sub>2</sub>, CO, CO<sub>2</sub>, N<sub>2</sub> and H<sub>2</sub>O present. (These gases were also detected by Heller and Gordon<sup>18</sup> and by Lengelle, et al.)<sup>19</sup> Previous documentation of the catalytic effect of platinum on both the water-gas and hydrogen-nitric oxide reactions, led them to test for these effects on their Pt-Pt/Rh thermocouples. To do this, they flowed a hot mixture of 5% H<sub>2</sub> in NO over bare and borax-coated Pt-Pt/Rh thermocouples. The coated thermocouple read 1034°C compared with 1416°C for the uncoated one. (These findings have been corroborated by Thomas and Freeze.)<sup>20</sup> Borax coating also had a dramatic effect on the solid propellant (M-2 double base) temperature profile. At 150 psig, with uncoated thermocouples, Klein, et. al.,<sup>7</sup> observed a second temperature stage from 1100°C to 1733°C (melting point of Pt). This stage disappeared when coated thermocouples were used.

In view of the unambiguous results just described, it is surprising that subsequent investigators, who checked for catalytic effects with Pt-Pt/Rh thermocouples, found none. Heller and Gordon<sup>18</sup> and Hunt, Heller, and Gordon<sup>21</sup> tried various coatings including borax, alumina, and zirconium oxide. No significant differences in temperature between coated and uncoated couples were found, although they did note a higher mortality rate among the uncoated couples. Both small imbedded thermocouples (2.5 µm and 7.5 µm diameter wire) and larger surface-bearing thermocouples (75 µm) were used in this study. It is not clear from References 18 and 21 whether the negative finding pertained to the entire temperature profile or only to apparent surface temperatures (which did exceed 1100°C in some instances).

Sabadell, et al.,<sup>14</sup> tested for catalytic effects with their Pt-Pt/Rh thermocouples using a silica coating technique and found no significant differences. This study focused primarily on PBAA-AP composite propellant but did include a double base propellant. It is not clear whether the negative

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<sup>18</sup>C.A. Heller and A.S. Gordon, "Structure of the Gas Phase Combustion Region of a Solid Double Base Propellant," J. Phys. Chem., Vol. 59, pp. 773-777, 1955.

<sup>19</sup>G. Lengelle, J. Duterque, C. Verdier, A. Rizot, and J. Trubert, "Combustion Mechanisms of Double Base Solid Propellants," 17th Symposium (International) on Combustion, The Combustion Institute, p. 1443, 1978.

<sup>20</sup>D.B. Thomas and P.D. Freeze, "The Effects of Catalysis in Measuring the Temperature of Incompletely-Burned Gases with Noble-Metal Thermocouples," Temperature: Its Measurement and Control in Science and Industry, Ed. by Harmon H. Plumb, Instrument Society of America, Part 3, Vol. 4, pp. 1671-1676, 1972.

<sup>21</sup>M.H. Hunt, C.A. Heller, and A.S. Gordon, "Surface Temperatures of Burning Double Base Propellants," NAVORD Report 2079, China Lake, CA, 1954.



test result included one or both of these propellants. Measured temperatures did exceed 1100°C with both propellant types.

Rogers and Suh<sup>22</sup> used surface-bearing thermocouples to measure surface temperatures in M-2 double base propellant. Both Pt-Pt/Rh and Chromel-Alumel couples gave the same results suggesting no catalytic activity in the 300-450°C range. Suh, et al.,<sup>17,23</sup> also used imbedded thermocouples of both types (also in M-2 propellant) and reported no differences. Their maximum pressure was 100 psig and their maximum temperatures reached were less than 1100°C. Thus, the threshold for catalytic activity may not have been reached.

On the strength of the Klein, et al.,<sup>7</sup> findings, one may conclude that, for double base propellants, at least, catalytic effects are likely to cause serious error in temperature measurements above 1100°C made with bare Pt-Pt/Rh thermocouples. A number of coatings have been used, which appear to eliminate these effects. These coatings, however, may aggravate the response problem.

The above discussion, refers to studies utilizing Pt-Pt/Rh thermoelements, which are inherently unreactive materials. Zenin<sup>15</sup> used imbedded ribbon thermocouples constructed of tungsten-rhenium alloys to map temperature profiles in double-base propellants through the entire combustion wave. These measurements included the visible flame zone where temperatures exceeded 2000°C. The desirability of such broad temperature response is obvious, but the possibility exists that the measurements are effected by reaction between the tungsten and incompletely burned gases. Above 500°C tungsten oxidizes rapidly in an oxygen environment<sup>24</sup> and, presumably, even more rapidly in the presence of NO<sub>2</sub>, a probable fizz zone component. It may be that this reaction is slow compared to the residence time of the thermocouple in the flame zone, but investigation of the matter would be prudent since thermoelement oxidation can lead to calibration shifts.

#### F. Solid Phase Response

In the discussion to follow, we apply the term "response" to the ability of the thermocouple bead to track the true temperature profile existing in the combustion wave. In the solid, this characteristic depends on several interrelated factors. The volumetric heat capacity of the thermocouple junction (for Pt, Pt 10% Rh) is about 50% higher than for double base propellant. Thus the junction, which is subjected to the same heat input as the surrounding propellant, will tend to lag the temperature of the

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<sup>22</sup>C.R. Rogers and N.P. Suh, "Ignition and Surface Temperatures of Double Base Propellants at Low Pressures II. Comparison of Optical and Thermocouple Techniques," AIAA J., Vol. 8, pp. 1501-1506, 1970.

<sup>23</sup>C.L. Thompson Jr. and N.P. Suh, "Gas Phase Reactions near the Solid-Gas Interface of a Deflagrating Double-Base Propellant Strand," AIAA J., Vol. 9, pp. 154-159, 1971.

<sup>24</sup>F.R. Caldwell, "Thermocouple Materials," in Temperature: Its Measurement and Control in Science and Industry, Reinhold Publishing Corp., Vol. 3, Part 2, pp. 81-134, 1962.

propellant. On the other hand, the thermal conductivity of the thermocouple material is about three orders of magnitude larger than that of the propellant. This means that the temperature within the bead will be essentially uniform. Because of the finite size of the junction, this results in perturbations to the temperature field one is trying to measure. Since heat is drawn from the propellant in front of the bead and deposited into the propellant behind the bead, the measured temperature at some instant may even be higher than would exist in the absence of the bead. Coupled to these factors is the effect of heat conduction through the thermocouple leads, which probably tends to depress the measured temperature from the true value. The most notable attempt to calculate the combined effects of these influences was made by Suh and Tsai<sup>11</sup> which we discussed briefly above. An adequate treatment of the problem would involve numerical solution of the 3-D, transient heat conduction problem, a formidable research task in itself, given the steepness of the temperature gradients expected.

Instead of addressing the full 3-D geometry, we have chosen to solve a simpler problem which, nonetheless, can be expected to yield insights to the general nature of the thermocouple response in the solid phase. Our problem consists of a thin slab of thermocouple material imbedded in a semi-infinite slab of propellant burning steadily at constant pressure. This involves solving only the 1-D, transient heat conduction equations and may be viewed as an idealization of a ribbon thermocouple (Zenin)<sup>15</sup> whose leads are directed away from the junction along an isotherm for a sufficient distance to eliminate the lead conduction part of the problem.

Mathematical details of the solution are given in Appendix A, but a few comments are made here regarding how the problem was set up. First, we assume that any condensed phase heat release occurs in a vanishingly thin zone at the surface. The undisturbed temperature profiles are then simple exponential functions and based on the surface temperatures reported by Kubota.<sup>9</sup> The surface boundary condition for the thermocouple-disturbed case is taken to be the heat flux into the solid that exists for the undisturbed case. The reasoning here is that the gas phase heat feedback varies only slightly for small changes in the surface temperature, which might be caused by the thermocouple slab. However, when the thermocouple plate emerges from the surface, the heat feedback should drop to zero since gas phase reactants are no longer being produced. Since this behavior is a consequence of our thermocouple slab being of infinite extent, we chose to terminate the calculation when the leading edge of the thermocouple slab is within 1 micron of the surface. This defect prevents us from determining exact corrections to the measured surface temperature but still allows trends to emerge.

The disturbance to the temperature field, caused by the thermocouple slab, is illustrated in Figures 1-3. These plots show the instantaneous temperature distribution (dashed line) in the solid when the leading edge of the thermocouple slab is five microns from the burning propellant surface. The temperature profile (solid line) in the absence of the thermocouple, is also shown for comparison. Figure 1 shows the comparison when the burning rate is 0.05 cm/sec (1.2 atm). The slight disturbance of the profile behind the thermocouple plate indicates that the specific heat effects are probably dominating the response. At the higher burning rate of .32 cm/sec (21 atm) shown in Figure 2 for the same plate thickness, considerably more disturbance is seen behind the thermocouple. Dynamic effects due to the high thermal



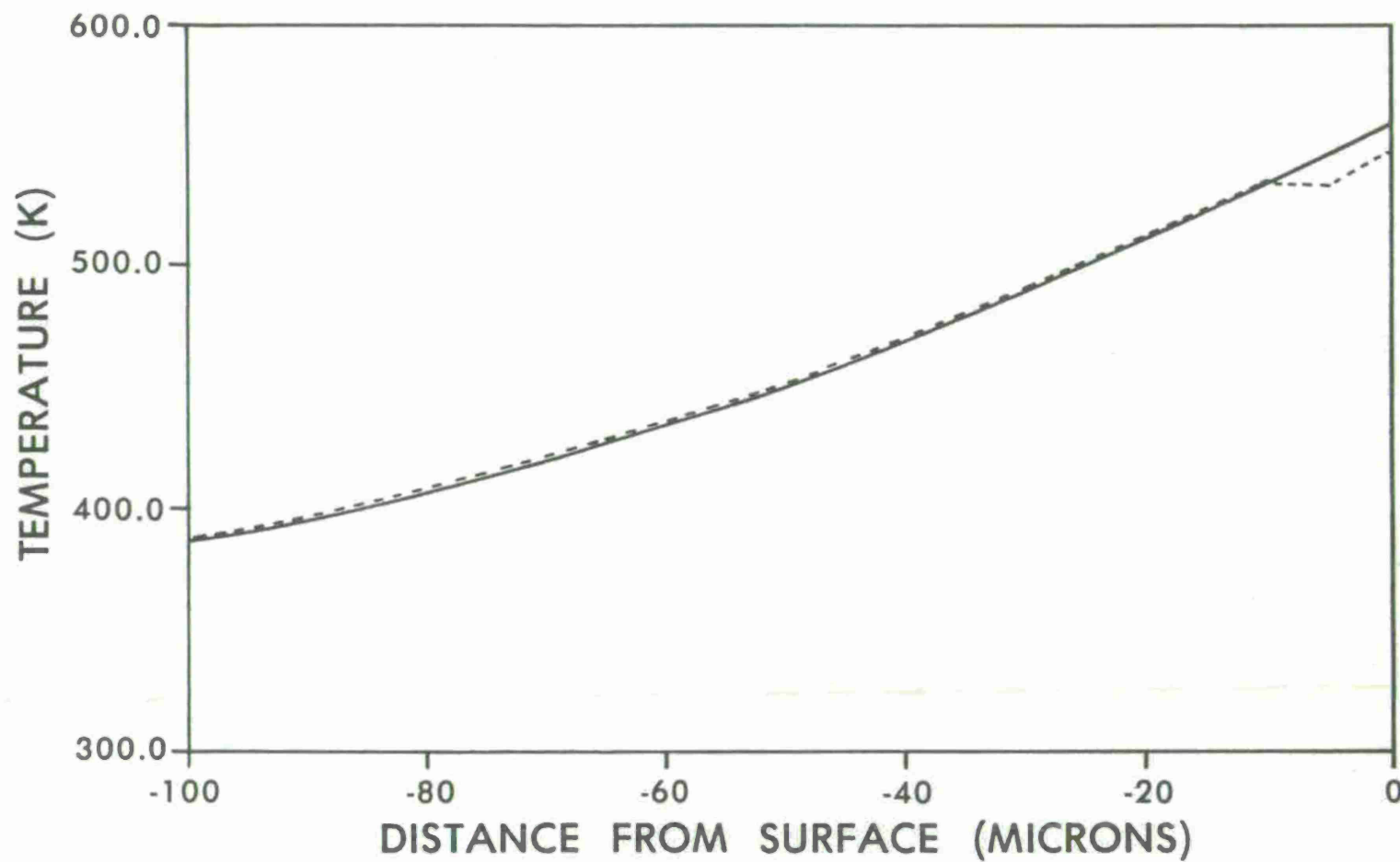


Figure 1. Instantaneous Temperature Field Disturbance (Dashed Line) In The Condensed Phase Compared With The Undisturbed Profile (Solid Line).  $P=1.2$  atm,  $d=5$   $\mu\text{m}$ .

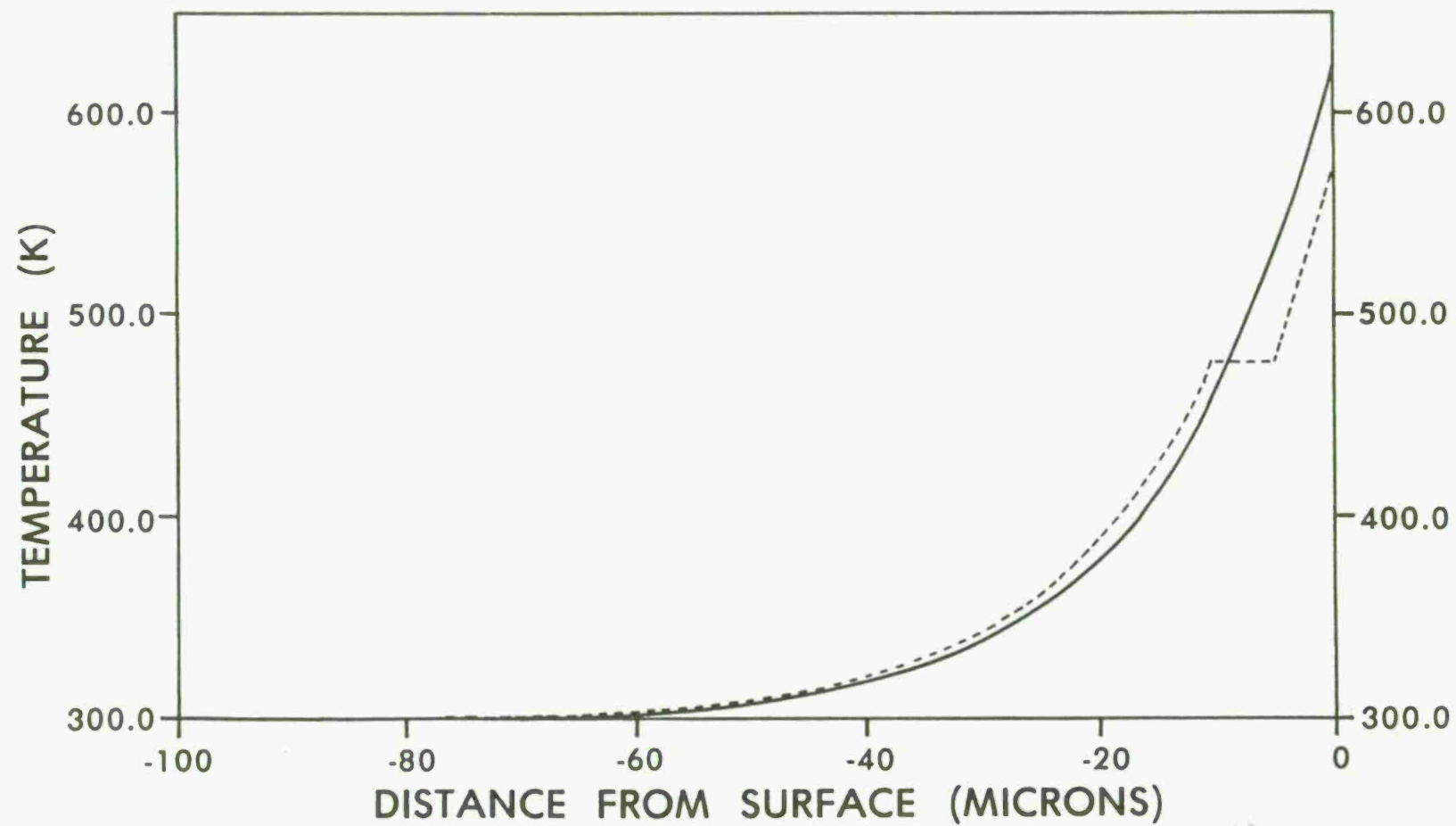


Figure 2. As In Figure 1, Except  $P=21$  atm,  $d=5$   $\mu\text{m}$ .

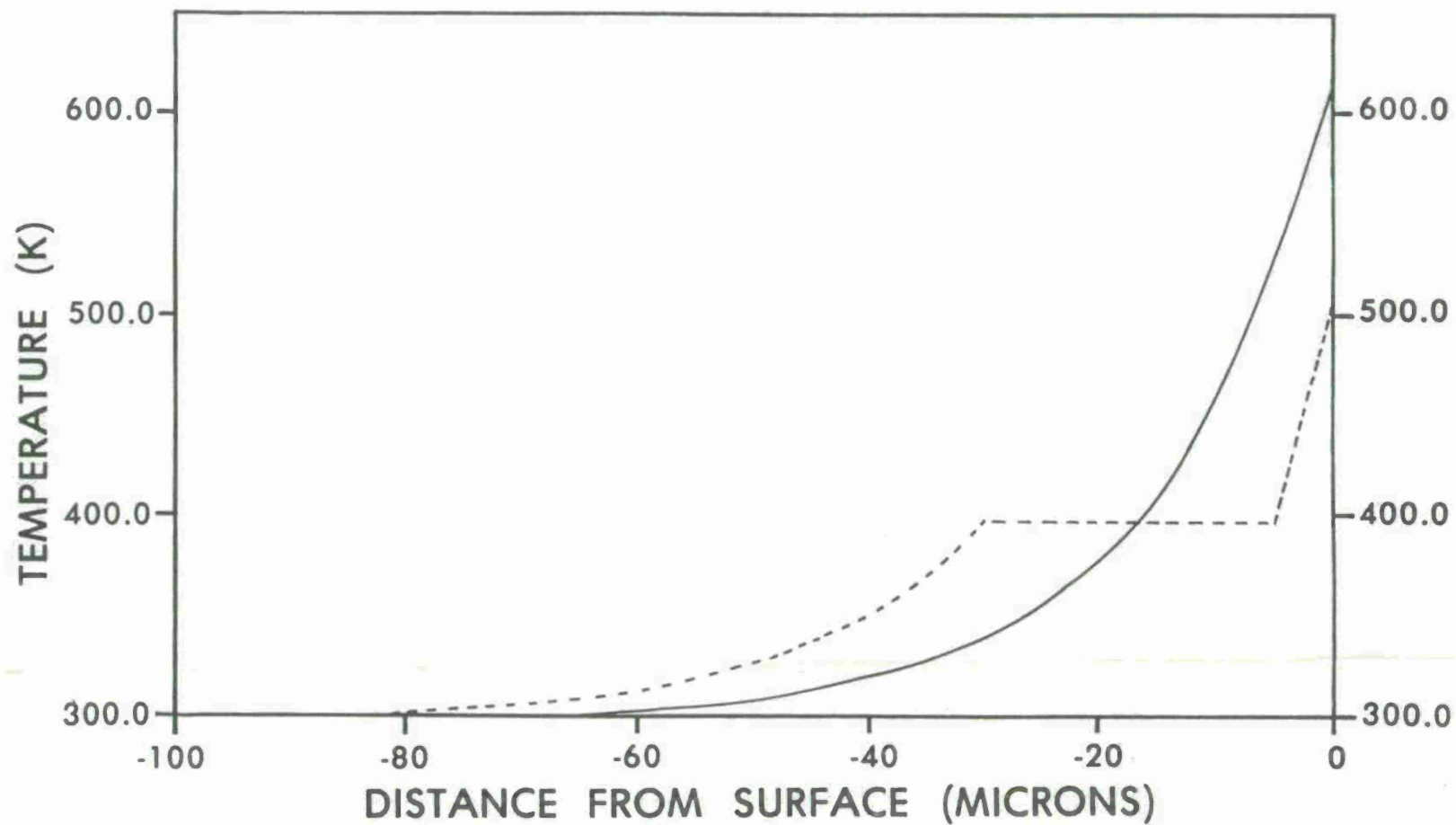


Figure 3. As In Figure 1, Except  $P=21$  atm,  $d=25$   $\mu\text{m}$ .

conductivity of the plate, are evidently assuming greater importance. Figure 3 illustrates the dramatic distortions of the temperature profile by a large (25 micron) thermocouple plate at the higher burning rate.

The errors in temperature profile measurements are shown in Figures 4-6, where the difference between the thermocouple plate temperature and the undisturbed temperature (at the center of the plate) is plotted as a function of the distance from plate center to the propellant surface. Note that at the highest burning rate the larger thermocouples may give smaller errors over a portion of the profile (although not near the surface). In these cases, the disturbance to the profile caused by the relative conductivity effects compensates for the lags due to the relative specific heats. Unfortunately, such compensation is not uniform enough over the entire profile to prove useful. In Figure 6, it is seen that the dynamic effects become so large for the 25 micron plate that the thermocouple reads a higher temperature than is actually present over at least a portion of the temperature profile.

The ribbon thermocouples used by Zenin<sup>15</sup> were 3.5  $\mu\text{m}$  thick by 50  $\mu\text{m}$  wide. Using Figure 6 one would estimate that a 3.5  $\mu\text{m}$  thick plate might underestimate the surface temperature by  $\sim 35^\circ\text{C}$ . For the 4  $\mu\text{m}$  diameter bead thermocouples used by Kubota,<sup>9</sup> one might expect considerably lower errors in surface temperature due to the much greater lateral surface area which enhances thermal accommodation. Unfortunately, the lead conduction problem will tend to exacerbate the temperature lags to a rather unpredictable extent. Ultimately, one must determine these temperature errors by examining a sequence of temperature profiles using successively smaller thermocouples.

#### G. Gas Phase Response

Unlike the solid phase, response errors in the gas phase have been estimated by a number of researchers.<sup>7,15,25</sup> In each case, the theory of convective heat transfer was used to determine the rate of heating of the thermocouple as it passes through the fizz and dark zones. Each also corrected for heat loss from the thermocouple by radiation. These corrections are expressed by the equation

$$c_B \rho_B V_B \frac{dT_B}{dt} = hA_B (T_G - T_B) - \sigma \epsilon A_B T_B^4 \quad (3)$$

where  $c_B$ ,  $\rho_B$ ,  $V_B$ ,  $A_B$ , and  $T_B$  are the specific heat, density, volume, surface area, and temperature, respectively, of the thermocouple bead.  $T_G$  is the undisturbed gas temperature,  $h$  the heat transfer coefficient,  $\sigma$  the Stefan-Boltzman constant, and  $\epsilon$  the emissivity of the bead. We assume the radiation heat gain by the thermocouple is negligible, thereby maximizing the loss. In addition, the leads from the junction are assumed to be arranged along an isotherm through the bead so as to eliminate the lead conduction complication.

The heat transfer coefficient is given by

<sup>25</sup>V.E. Zarko, A.I. Sukhinin, and S.S. Khlevnoi, "Temperature Measurement in the Gas Phase of a Burning Propellant," Fizika Goreniya i Vzryva, Vol. 3, pp. 45-53, 1967.

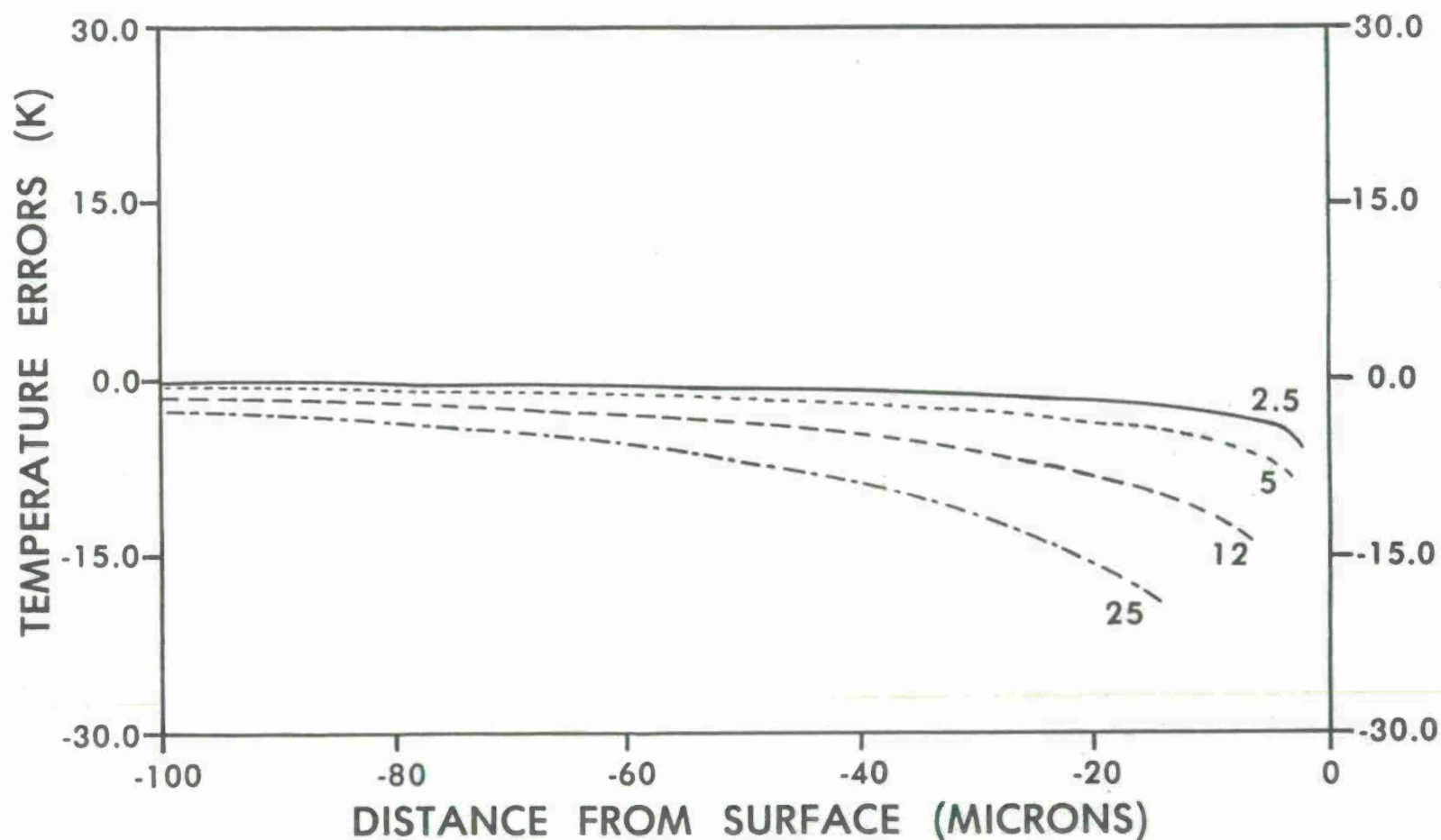


Figure 4. Condensed Phase Response Errors: Difference Between Undisturbed Temperature At Center Of Plate And Thermocouple Plate Temperature.  $d=2.5 \mu\text{m}$ ,  $5 \mu\text{m}$ ,  $12 \mu\text{m}$ , And  $25 \mu\text{m}$ .  $P=1.2 \text{ atm}$ .

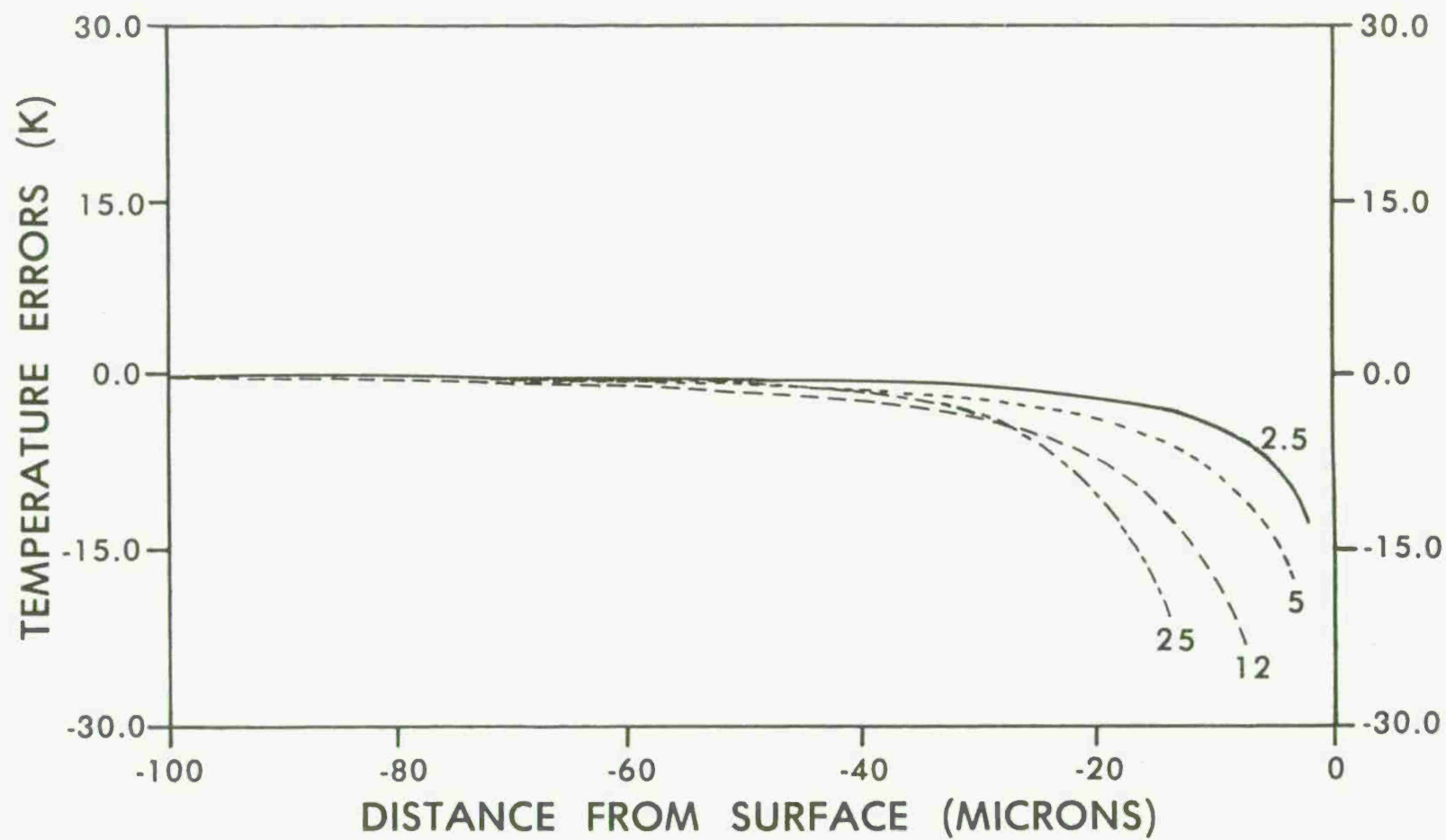


Figure 5. As In Figure 4, Except  $P=8$  atm.



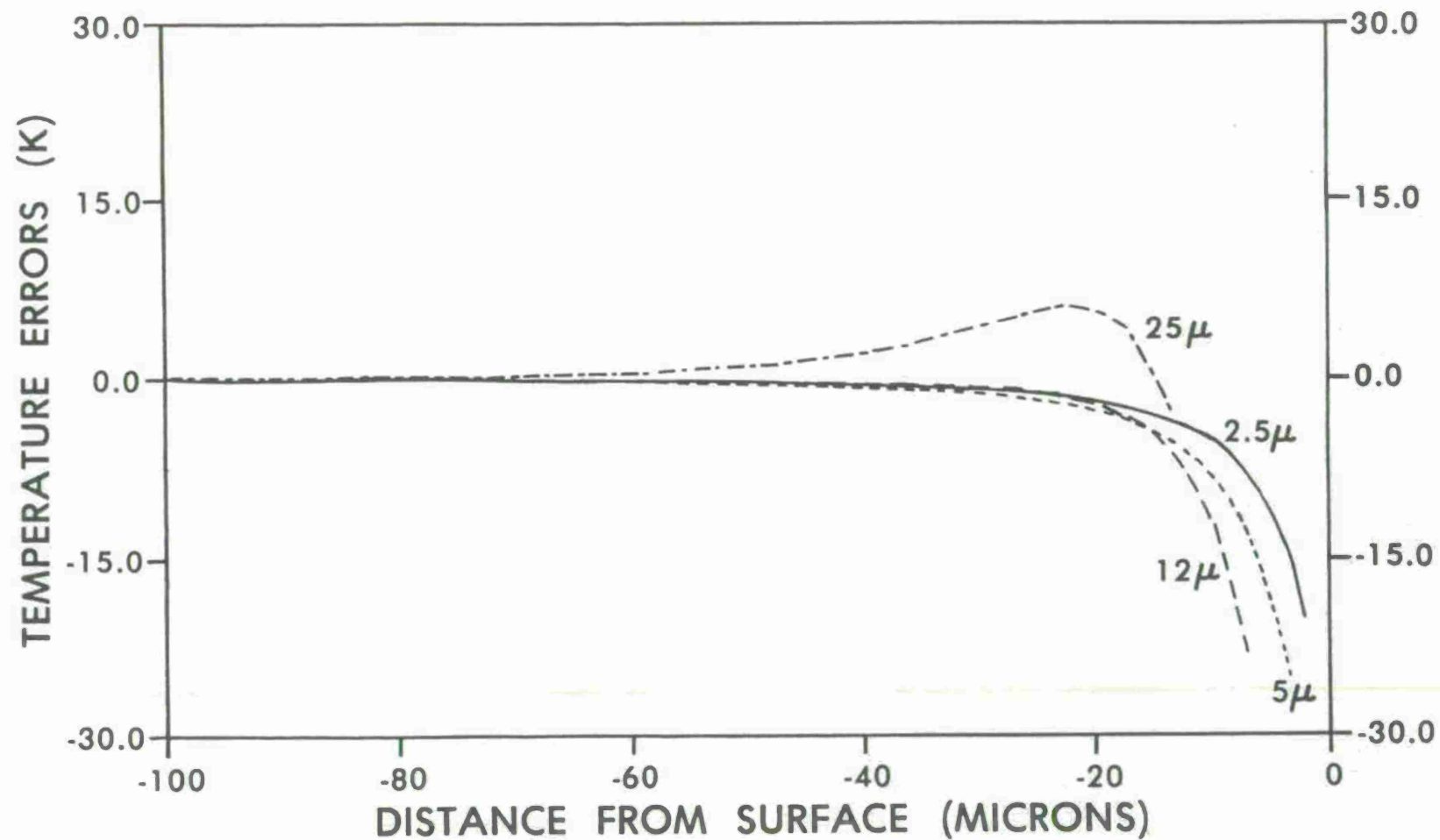


Figure 6. As In Figure 4, Except P=21 atm.

$$h = Nu \frac{\lambda}{d} \quad (4)$$

where Nu is the Nusselt number,  $\lambda$  the gas thermal conductivity and d the bead diameter. The Nusselt number has been empirically correlated with the Reynolds number by Hilpert<sup>26</sup> for Reynolds numbers in the range 1 to 4. Ulsamer<sup>26</sup> gives about the same values down to Re = .1.

$$Nu = .89 Re^{.33} \quad (5)$$

This correlation pertains to flow over cylinders. For spheres at much higher Reynolds numbers, Williams<sup>26</sup> gives a correlation which has about the same exponent as cylinders at those Re but has about twice the coefficient. Thus the above correlation is probably a bit low, which will lead to overestimates in the temperature lags (compensating, at least in part, for our neglect of lead conduction). Values of the other parameters are given in Appendix A.

Figure 7 shows the above correction applied to the measured temperature profile of Kubota, et. al.,<sup>9</sup> for a 10 micron bead diameter at 21 atm. This measurement was actually made with a bead diameter of about 4 microns, but the correction is slight and better illustrated by the 10 micron assumption. In Table 1 the estimated maximum temperature lags are given for other pressures and bead sizes. This table is based upon the use of Kubota's<sup>9</sup> temperature profiles as the measured ones, i.e.,  $T_B$  in Eq. (3). This procedure is not strictly consistent as one should first correct the measured profile using a 4 micron bead, then integrate Eq. (3) using the corrected profile as  $T_G$ . Our abbreviated method should overestimate the lag, and at least for the smaller errors, give approximately the same values.

TABLE 1. ESTIMATED GAS PHASE RESPONSE ERRORS

<u>P (atm)</u>	<u>Bead Diameter (microns)</u>	<u>Max Temp. Lag (K)</u>	<u>Max. Error due to Rad. Loss (K)</u>
1.2	40	21	2.1
	20	6.5	1.2
	10	2.0	0.8
	5	0.8	0.5
8	40	80	4.0
	20	25	2.3
	10	8	1.5
	5	3	0.9
21	40	300	15.8
	20	88	9.4
	10	28	5.7
	5	8	3.5

<sup>26</sup>As quoted in Jakob, Max, Heat Transfer, Vol. 1, pp. 560-564, 1949.



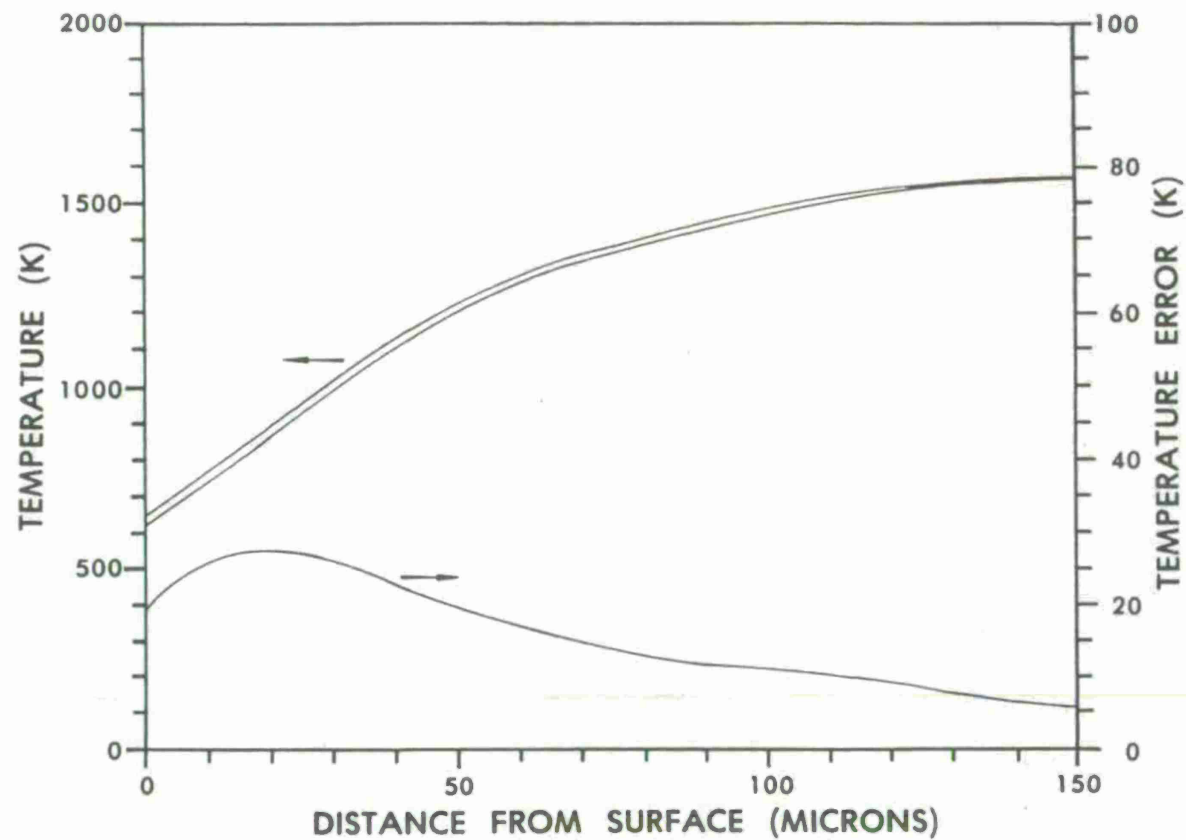


Figure 7. Corrected (via Eq. 3), Uncorrected Temperature Profiles And Net Errors In The Gas Phase.  $P=21$  atm, Bead Diameter = 10  $\mu\text{m}$ .

Although application of the Eq. (3) is straightforward, the validity of the heat transfer coefficient approach is not assured. It is based upon transient heat transfer to a body in a constant temperature gas stream where the temperature far from the boundary layer is the same in all directions. Its accuracy in the present case, involving a time varying steep gradient, has not been established.

Some perspective as to the sensitivity of the derived kinetics to these systematic errors can be gained by applying the analysis of Reference 6 to the corrected and uncorrected profiles in Figure 7. The effective activation energy and reaction order for the uncorrected profile are 5.86 kcal/mole and 1.10, respectively. The corrected profile produces values of 5.77 kcal/mole and 1.08. Thus, at least in this instance, a maximum error of some 30°C does not interfere significantly with the ultimate purpose of the measurement.

### III. CONCLUSIONS

Recent theoretical work has provided a firm base upon which to construct a practical description of propellant combustion. The old notion of using a single overall reaction to represent the fizz zone kinetics has been shown to be sound. However, important provisos are attached to this finding, viz., the kinetics parameters for this single reaction are not likely to result from a rate-limiting step and these parameters can be expected to vary with pressure and possibly initial temperature. Fortunately, a method has been developed<sup>6</sup> to obtain these effective kinetics values from a single temperature profile measured through the fizz zone of a burning propellant. Since the imbedded thermocouple method is the only technique presently capable of providing this critical data, this report has attempted to identify and assess the errors likely to interfere with the accuracy of such a measurement.

Although such an effort seemed prudent before embarking upon a costly and tedious experimental program, our findings have not completely removed the risk from such an undertaking. A complete mathematical treatment of the thermocouple errors in the condensed and gas phases is clearly not the most expedient approach to evaluating the technique. However, since all of the errors diminish with thermocouple size, a systematic experimental effort using a progression of sizes should be sufficient to evaluate the adequacy of microthermocouple measurements. The analysis for the effective kinetics, in fact, provides a means for judging this adequacy in relation to the intended purpose of the data.

Notwithstanding the qualifications attached to our analyses here, a few conclusions can be drawn which give a fair assessment to the imbedded thermocouple method and guidance for its implementation.

1. In order to be able to judge the convergence of temperature profiles with diminishing thermocouple size, one should strive for exacting control of junction fabrication and burning conditions (pressure, initial temperature, inhibition, flow, etc.).
2. Temperature errors due to catalytic effects are probably not a problem below 1100°C. Above this temperature, coating may be required but may exacerbate the response error problem.

3. There may be an optimum angle between the leads of the junction, minimizing heat loss from the bead through the leads on the one hand, and maximizing the mechanical stability of the junction on the other. This possibility should be studied experimentally.

4. The surface temperature is an important feature of the temperature profile and Zenin's electrical conductivity measurement, or an extension of this idea is probably the most promising approach. For example, one might be able to sense the capacitance change between the bead and a thin wire a few microns closer to the surface as the wire is blown off the surface by the advancing combustion front.

5. A beaded thermocouple is probably superior in response to a ribbon thermocouple of comparable size due to enhanced lateral thermal accommodation and greater spatial resolution should the propellant surface not burn exactly parallel to the plane of the ribbon.

6. Estimates of gas phase response errors suggest temperature lags of about 10K for a thermocouple bead diameter of 5 microns in the case of a double base propellant burning at 21 atm (3.2 mm/s).

A final comment is appropriate on the importance of these temperature measurements to the broader problem of interior ballistics. The implication of this review is that microthermocouples can likely be used to deduce the amount and effective rate of the heat release for solid propellants up to about 20 atm. The experiments suggested may show the method to be valid at somewhat higher pressures as well, but meaningful results are not likely to be obtained above about 50 atm. Since during the ballistic cycle pressures of thousands of atm are reached, one may legitimately question the value of these low pressure diagnostics. Typically, nitrate ester propellants change from flameless fizz burning to full flame burning in the pressure range 7-15 atm. In this transition, the total heat release approximately doubles. Recent research using ballistic simulators<sup>27</sup> and interior ballistics codes<sup>28</sup> has suggested that a quantitative description of this transition phenomena may be critical to successful model predictions of the early ignition sequence in guns. The imbedded thermocouple technique with ancillary analysis<sup>6</sup> may provide a means to this end.

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<sup>27</sup>Thomas C. Minor, "Experimental Studies of Multidimensional Two-Phase Flow Processes in Interior Ballistics," U.S. Army Ballistic Research Laboratory Report ARBRL-MR-03248, April 1983. (AD A128 034)

<sup>28</sup>K.D. Fickie and G.E. Keller, "An Investigation of Kinetics in Solid Propellant Ignition," Proceedings of the 21st JANNAF Combustion Meeting, October 1984.



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APPENDIX A  
CONDENSED PHASE RESPONSE SIMULATION





# APPENDIX A

## CONDENSED PHASE RESPONSE SIMULATION

The one-dimensional transient heat conduction equation for constant thermal conductivities ( $\lambda_B$  in the thermocouple and  $\lambda$  in the propellant) and specific heats ( $c_B$  and  $c$ ) may be written

$$\frac{\partial T}{\partial t} = \frac{\lambda}{\rho c} \frac{\partial^2 T}{\partial x^2} - r \frac{\partial T}{\partial x} \quad (A1)$$

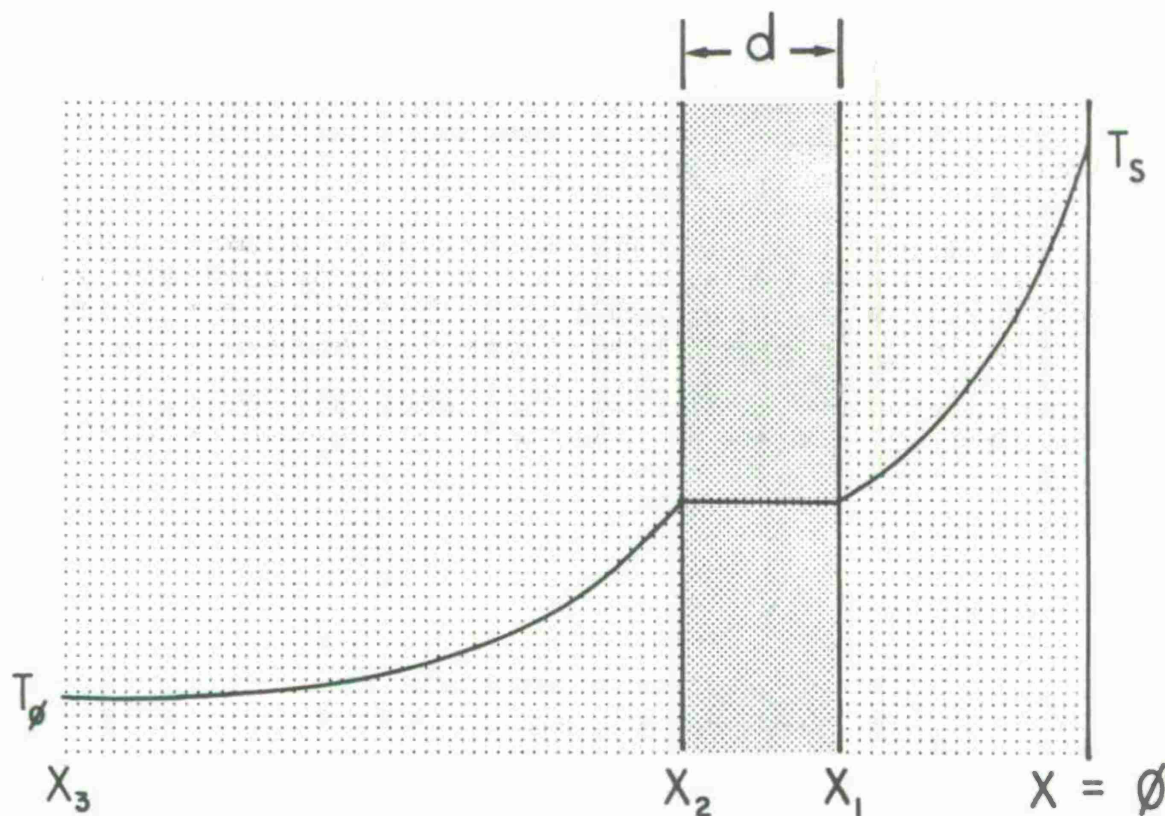


Figure A-1. Geometry For Calculation Of Condensed Phase Thermocouple Response.

Figure A-1 shows the geometry of the thermocouple slab in relation to the burning surface. We locate the propellant surface at  $x=0$ , the right side of the thermocouple at  $x_1$ , and the left side of the thermocouple at  $x_2$ . The left

boundary theoretically should be at  $x=-\infty$ , but instead we choose a value  $x_3$  small enough so there is no appreciable temperature gradient. The boundary conditions are

$$\begin{aligned}
 x=x_3 \quad T &= T_0 = 298K \\
 x=x_2 \quad T(x_2^-) &= T(x_2^+) \\
 \lambda \frac{\partial T}{\partial x} \Big|_{x_2^-} &= \lambda_B \frac{\partial T}{\partial x} \Big|_{x_2^+} \\
 x=x_1 \quad T(x_1^-) &= T(x_1^+) \\
 \lambda_B \frac{\partial T}{\partial x} \Big|_{x_1^-} &= \lambda \frac{\partial T}{\partial x} \Big|_{x_1^+} \\
 x=0 \quad \lambda \frac{\partial T}{\partial x} \Big|_{0^-} &= \rho r c (T_s^0 - T_0) \quad . \quad (A2)
 \end{aligned}$$

As an initial condition, the thermocouple is far from the burning surface. The temperature profile at  $t=0$  is the analytic solution to the problem of a thermocouple plate remaining at a fixed distance from the surface. This profile is essentially that found in the absence of a thermocouple.

To solve the transient equation, we first perform a change of coordinates. Consider first the hot propellant region between the surface and thermocouple. Let

$$E_1(t) = -x_1 \quad (A3)$$

be the size of this interval. Then

$$E_1(t) = E_1(0) -rt \quad (A4)$$

by the definition of the regression rate and

$$\frac{dE_1}{dt} = -r \quad . \quad (A5)$$

Now consider new coordinates  $x'$  and  $t'$  defined by

$$x=x'E_1 \quad , \quad t=t'. \quad (A6)$$

Note that  $-1 \leq x' \leq 0$  for all times  $t$ . Then

$$\frac{\partial T}{\partial x'} = \frac{\partial T}{\partial x} \frac{\partial x}{\partial x'} + \frac{\partial T}{\partial t} \frac{\partial t}{\partial x'} \quad (A7)$$

$$= E_1 \frac{\partial T}{\partial x}$$

$$\frac{\partial^2 T}{\partial x'^2} = E_1^2 \frac{\partial^2 T}{\partial x^2} \quad (A8)$$

$$\begin{aligned}\frac{\partial T}{\partial t'} &= \frac{\partial T}{\partial x} \frac{\partial x}{\partial t'} + \frac{\partial T}{\partial t} \frac{\partial t}{\partial t'} \\ &= -x'r \frac{\partial T}{\partial x} + \frac{\partial T}{\partial t}\end{aligned}\quad (A9)$$

or substituting into (A1),

$$\frac{\partial T}{\partial t'} = \frac{\lambda}{\rho c} \frac{1}{E_1^2} \frac{\partial^2 T}{\partial x'^2} - (rx' + r) \frac{1}{E_1} \frac{\partial T}{\partial x'} \quad (A10)$$

Similarly, for the thermocouple region, we let

$$E_2 = x_1 - x_2 \quad (A11)$$

and

$$x = E_1 + x' E_2, \quad t = t' \quad (A12)$$

Again,  $-1 \leq x' \leq 0$ . In this case

$$\frac{\partial T}{\partial x'} = E_2 \frac{\partial T}{\partial x} \quad (A13)$$

$$\frac{\partial^2 T}{\partial x'^2} = E_2^2 \frac{\partial^2 T}{\partial x^2} \quad (A14)$$

$$\begin{aligned}\frac{\partial T}{\partial t'} &= \frac{\partial T}{\partial t} + \frac{\partial T}{\partial x} \frac{\partial x}{\partial t'} \\ &= \frac{\partial T}{\partial t} - \frac{r}{E_2} \frac{\partial T}{\partial x'}\end{aligned}\quad (A15)$$

$$\text{So, } \frac{\partial T}{\partial t'} = \frac{\lambda_B}{\rho c_B} \frac{1}{E_2^2} \frac{\partial^2 T}{\partial x'^2} \quad (A16)$$

The derivation for the colder propellant region is analogous and results in

$$\frac{\partial T}{\partial t'} = \frac{\lambda}{\rho c} \frac{1}{E_3^2} \frac{\partial^2 T}{\partial x'^2} \quad (A17)$$

where  $E_3 = x_3 - x_2$ ,  $x = E_1 + E_2 + x' E_3$ , and  $t = t'$ .

The transformation of the boundary conditions is straightforward. We now have a set of three partial differential equations over a fixed internal  $-1 \leq x' \leq 0$ . The equations are solved using PDECOL,\* a package for the solution of partial differential equations.

---

\*N.K. Madsen and R.F. Sincovec, "PDECOL: General Collocation Software for Partial Differential Equations," Preprint UCRL-78263 (rev 1), Lawrence Livermore Laboratory, 1977.

There is one additional problem to be overcome. PDECOL was designed to solve a set of partial differential equations over the same interval. Because of our coordinate transformation, all three equations can be viewed as being defined on the interval  $-1$  to  $0$ . However, it is also assumed that the boundary conditions are equations involving only quantities defined at the left end of the interval ( $x' = -1$ ) or at the right end ( $x' = 0$ ). But the boundary conditions for the thermocouple involve both ends of the interval.

The solution is to introduce a new coordinate for the thermocouple region,  $x'' = -x'$ . The only effect is that the hot side of the thermocouple is now  $x'' = -1$  and the cold side is  $x'' = 0$ . We now have three boundary conditions at  $0$ , that is, at  $x=0$  and at  $x=x_2$ ; and three conditions at  $-1$ , that is, at  $x=x_3$  and  $x=x_1$ . The numerical solution is now straightforward.

TABLE A-1. PHYSICAL PROPERTIES

Property	Symbol	Value
Thermal conductivity of thermocouple material (Pt)	$\lambda_B$	.18 cal/cm-s-K
Density of thermocouple material (Pt)	$\rho_B$	21.5 g/cm <sup>3</sup>
Specific heat of thermocouple (Pt)	$c_B$	.033 cal/g-K
Thermal conductivity in propellant	$\lambda$	$2.5 \times 10^{-4}$ cal/cm-s-K
Density of propellant	$\rho$	1.6 g/cm <sup>3</sup>
Specific heat of propellant	$c$	.34 cal/g-K

TABLE A-2. UNDISTURBED TEMPERATURE PROFILE DATA\*

P (atm)	r (cm/s)	$T_s$ ° (K)
1.2	.05	560
8	.158	576
21	.321	618

\*Based on experimental work of Kubota, et al.<sup>9</sup>



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